Comparison of Carbonyls and BTEX Emissions from a Light Duty Vehicle Fuelled with Gasoline and Ethanol-Gasoline Blend, and Operated without 3-Way Catalytic Converter

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ABSTRACT

This paper presents the comparison of unregulated emissions such as carbonyls and BTEX (Benzene, Toluene, Ethyl Benzene, and Xylenes) species emanated from a light duty SI (Spark Ignition) vehicle E-0 (fuelled on gasoline) and E-10 (ethanol-gasoline blend). Meanwhile, the ozone forming potential of these pollutants based on their ozone SR (Specific Reactivity) has also been addressed in this study. The experiments were performed on transient as well as steady-state modes in accordance with the standard protocols recommended for light duty vehicle emissions. Carbonyls and BTEX were analyzed by HPLC (High Performance Liquid Chromatography) with UV detector and GC/MS (Gas Chromatography/Mass Spectroscopy), respectively. Formaldehyde and acetaldehyde were the predominant components of the carbonyls for E-0 and E-10, respectively. During transient mode, formaldehyde, acrolein + acetone, and tolualdehyde pollutants were decreased but, acetaldehyde emissions increased with E-10 as compared to E-0. The BTEX emissions were also decreased with E-10, relative to E-0. During the steady-state modes, formaldehyde, acrolein + acetone and propionaldehyde were lower, aromatic aldehydes were absent, but acetaldehyde pollutants were higher with E-10 compared to E-0. The BTEX emissions were decreased at medium and higher speed modes however, increased at lower speed mode with E-10 as compared to E-0. Total BTEX emissions were maximal at lower speed mode but, least at medium speed mode for both the fuels. SR of the pollutants was higher over transient cycle of operation, compared with steady-state mode. Relative to E-0, E-10 displayed lower SR during both transient as well as steady-state mode.

Key Words: Alternative Fuel, Ethanol, Unregulated Emissions, Carbonyls, Volatile Organic Compounds.

1.   INTRODUCTION

The unrelenting use of fossil fuels and its consequences on human life and environment are instigating the scientists and researchers to endeavor for the alternative sources of energy. Among the number of alternative fuels, bio-fuels such as methanol, ethanol, and vegetable oils and their esters are receiving increasing attentions during the past few years, owing to their oxygenated, biodegradable and renewable properties, and reduced discharge of carbon dioxide (CO_2).

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Ethanol is a promising alternative fuel for vehicles, which does not only minimize the dependence on imported petroleum fuels but also has the least adverse impacts on public health and environment. The molecules of ethanol contain a hydroxyl group (-OH) bounded to a carbon atom, thus its oxygen content favors the further combustion of gasoline being used as gasoline-ethanol blends [1]. It has high octane number and can be used as a substitute fuel without major engine modifications [2]. It has been reported that the use of gasohol, a mixture of 10% ethanol and 90% gasoline, in motor vehicles results in 5% decrease in THC (Total Hydrocarbon), 13% decrease in carbon monoxide (CO), and 5% increase in oxides of nitrogen (NO_x) emissions [3].

Recently, ethanol and gasoline are widely being used in blended forms in many countries like USA, Canada, Brazil, EU (European Union), and Thailand [4]. In the eastern Sweden, all the gasoline which is being sold in the market contains about 5% ethanol [2]. In China, nine provinces are using gasohol in the gasoline vehicles to reduce the CO and PM (Particulate Matter), and to decrease the import of fossil fuel [5]. The use of ethanol has been extended to CI (Compression Ignition) engine, and some studies have also been conducted on biodiesel-ethanol-diesel (BE-diesel), a new form of biofuel [5-7].

The main objective of the current work is to compare the carbonyls, BTEX species, and ozone forming potential of these pollutants emanating from an SI vehicle fuelled on gasoline and ethanol-gasoline blend, both in transient and steady-state modes. Although some literature has been reported on regulated emissions from ethanol-gasoline blended fuels, unregulated emissions, particularly carbonyls and BTEX emissions have not yet been addressed comprehensively. To best of authors' knowledge, so far no work has been reported for the ozone specific reactivity of such pollutants in case of ethanol-gasoline blend, using the Chinese transient cycle as well as steady-state modes. So, this study would be helpful to know the impact of ethanol on the emissions of carbonyls and BTEX components which are notorious for their human health hazard activities and global warming.

Carbonyls such as formaldehyde, acetaldehyde, acrolein + acetone, propionaldehyde and aromatic aldehydes (benzaldehyde and tolualdehyde) have been discussed here because of the two main reasons: First, these compounds are the major contributors to the total carbonyls. According to Pang, et. al. [5], formaldehyde, acetaldehyde, acrolein, and aromatic aldehydes account for above 82 and 85% of the total carbonyls with gasohol and gasoline, respectively. Second, compounds like formaldehyde, acetaldehyde, and acrolein have been reported as possible human carcinogens [8], acetaldehyde and acetone have been declared to be the major contributors to photochemical smog generation cycle [9].

A group of VOCs (Volatile Organic Compounds), commonly known as BTEX was also selected in this study because of their health hazard, carcinogenic, and ozone forming activities. Benzene and toluene are accused of cancer generation [9], and xylene isomers have the potential to convert significant amounts of $NO-NO_2$ [10], hence to increase the ground level ozone and deplete the stratospheric ozone.

2. EXPERIMENTAL SETUP

2.1 Test Vehicle, Fuels and Operating Conditions

The experiments were conducted on a 60 kW, 1.3 L, 4 cylinder and MPFI (Multi Port Fuel Injected) gasoline vehicle operating without a TWC (Three-Way Catalyst). The vehicle was run on a 1.0 m single roll

DC electrical chassis dynamometer (ONO SOKKI Inc.), in accordance with the cycle consisting of basic urban and extra urban parts as discussed elsewhere [11], however, shown here in Fig. 1. Basic urban part of the cycle consists of four sub-cycles which simulate the urban regions, and extra urban part of the cycle simulates the main motor-way with aggressive speed of the vehicle. The average speed of the test vehicle remained 33.58 km/h during the experiments. Moreover, the vehicle was also run at the speeds of 40, 80 and 120 km/h representing the steady-state modes 1, 2, and 3 respectively, for a period of 300 seconds. During the experiments, a separate fuel tank was used to avoid the interference which might occur due to the residues in oil pipeline, as shown in the experimental setup in Fig. 2.

Two test fuels were used in this study, which are unleaded gasoline having RON (Research Octane Number) 93 (E-0) and ethanol-gasoline blend containing 10% ethanol and 90% gasoline (v/v) (E-10). Gasoline was used as a baseline or reference fuel. The main properties of the test fuels are given in Table 1.
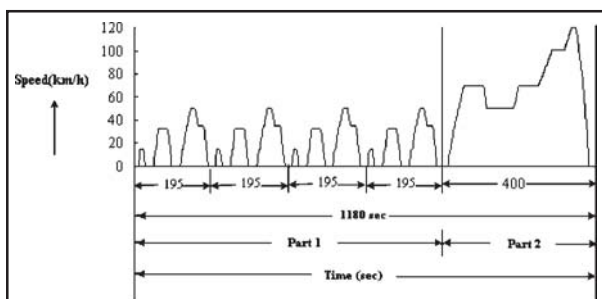

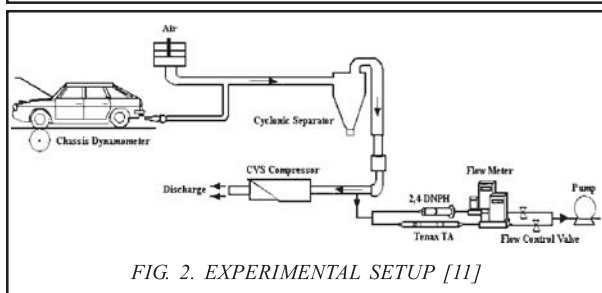*FIG. 1. OPERATING CYCLE OF TEST VEHICLE [11]*

FIG. 2. EXPERIMENTAL SETUP [11]

2.2   Sampling of Pollutants

The sampling scheme is shown in Fig. 2. In order to dilute the exhaust from the vehicle, a standard CFV-CVS (Critical Flow Venture-Constant Volume Sampling) dilution tunnel was used. The exhaust from the vehicle was mixed with the fresh and filtered ambient air at the temperature of about 25°C to dilute and cool the exhaust. The exhaust gas was cooled to ambient temperature which was maintained for all the tests to avoid the water condensation in the dilution tunnel. The exhaust flow rate was 10 m³/min and the dilution ratio was about 15. The exhaust samples were trapped in 2, 4-DNPH (Dinitrophenylhydrazine) coated silica gel cartridges (Accustandard® Inc.) and Tenax TA® tubes (Markes UK) for the carbonyls and BTEX emissions, respectively. The DNPH inside the cartridges reacted with the carbonyls, and hence formed the corresponding stable 2, 4-dinitrophenylhydrazone derivatives. The sampling was performed using a constant volume battery operated air pump (SKC USA, AirChek2000) at the flow rate of 220 mL/min for 10 min. Three samples of each E-0 and E-10 were taken for the carbonyls and BTEX emissions. After sampling, the tubes were refrigerated at about -10°C.

2.3   Sample Extraction and Analysis of Carbonyls

For the carbonyl species, the analytical method was based on the EPA (Environmental Protection Agency) method

TABLE 1. PROPERTIES OF FUELS

Properties	E-10	E-0
Gross Heat Content (MJ/kg)	42.2	46.0
Octane Number	95.0	93.0
Density (Kg/L) at 20°C	0.74	0.73
Carbonate Content (wt %)	83.4	86.4
Hydrogen Content (wt %)	13.1	13.6
Oxygen Content (wt %)	3.5	n/a

TO-11A [12].The carbonyls trapped sample was eluted by 3 mL acetonitrile on a SPE (Solid Phase Extractor), and the solution was collected in a 5 mL volumetric flask. The detailed procedure has already been discussed by authors elsewhere [11] however, is given here briefly for the interest. In order to analyze an aliquot of 25 μL, an HPLC system (Agilent 1200LC USA) was used, and the formed carbonyls-DNPH derivatives were separated on a column (Agilent Eclipse XDB C18, 4.6x150mm x 5μm). These species were then detected by a visible detector at 360 nm. Acetonitrile and water were used as mobile phases according to a volume ratio of 60 and 40% (v/v), respectively. The flow rate was 1.0 mL/min, and the temperature gradient was 25°C.

For the quantification of carbonyls compounds, an authentic commercial mixture (Supelco, USA) was used. This mixture contains 14 carbonyls which are FOR (Formaldehyde), ACE (Acetaldehyde), ACR (Acrolein), ATE (Acetone), PRO (Propionaldehyde), CRO (Crotonaldehyde), MEK (Methyl Ethyl Ketone), MET (Methacrolein), BUT (Butyraldehyde), BEN (Benzaldehyyde), VAL (Valeraldehyde), TOL (Tolualdehyde), CYC (Cyclohexanone), and HEX (Hexanaldehyde). The external standard methods were used to make linear standard curves of the compounds. The curves were calibrated using regression method in which correlation coefficient was more than 0.999. Since, it was difficult to resolve the chromatographic peaks of acrolein and acetone using an isocratic elution, so they were quantified together.

2.4 Sample Extraction and Analysis of BTEX Pollutants

For the BTEX species, the analytical method was based on the EPA standard method TO-17 [13]. The components trapped in Tenax TA® tubes were extracted using an automatic TD (Thermal Desorber), in which tubes were blown by the dry inert gases and then heated. The desorbed species were cryogenically focused in a cold trap at -10°C, and were heated to 280°C through a fused silica capillary column (HP-5MS, 30x0.25mm x 0.25μm) using GC (Gas Chromatography) (Agilent 6890N). The temperature program was 35°C (10 min) at 5°C/min to 280°C as discussed in detail elsewhere [14].

In order to identify and quantify the BTEX species, commercial standard solutions were purchased from Sino-Japan Friendship Center for Environmental Protection. The species were identified by comparing their retention times of chromatographic peaks, and hence comparing their mass spectra with those available in US NIST (National Institute of Standards and Technology) library. The BTEX components were quantified using external standard method as discussed above. In the MS (Mass Spectroscopy), the acquisition mode was SCAN, while the range of scan was 35-450 amu. Other properties of the MS were as under: electron multiplier voltage = 1.0 kV; Transfer line to MS temperature = 250°C; Ion source = EI (Electron Impact) 70 eV; Ion source temperature = 200°C; and Solvent cut time = 2.5 min.

3. RESULTS AND DISCUSSION

3.1 Transient Mode

3.1.1 Comparison of Carbonyls Emissions

Fig. 3(a) presents the EF (Emission Factor) of carbonyls emanated from the vehicle during transient mode of operation. It is clear that formaldehyde is the predominant component of carbonyls in case of E-0, whose RC (Relative Contribution) is above 55%. The next higher contributors are acetaldehyde, acrolein + acetone and propionaldehyde with their RC of 35, 30.8 and 10.7%, respectively. In case of E-10, acetaldehyde is the most abundant carbonyl with

RC of more than 58%. After acetaldehyde, next higher contributors are formaldehyde, propionaldehyde and acrolein + acetone with their RC of 34.4, 10.3 and 8.8%, respectively. This finding is in good agreement with those of previous studies that formaldehyde and acetaldehyde are two major aldehyde compounds in the exhaust of gasoline vehicles [11,15-16].

In case of E-10, there is no evidence of benzaldehyde emissions, and formaldehyde, acrolein + acetone and tolualdehyde decrease by 33, 68.5 and 79% respectively, compared with E-0. However, acetaldehyde emissions increase by 81% with E-10 as compared to E-0. This phenomenon of increase in acetaldehyde with decrease in formaldehyde with E-10 is also supported by other literature [4-5]. The increase in acetaldehyde in case of E-10 relative to E-0 may be attributed to the oxidation of ethanol to acetaldehyde. But, the emission of higher formaldehyde with E-0 is ascribed to the incomplete combustion of gasoline compared with oxygenated gasohol, as formaldehyde emitted from the vehicle exhaust is mainly due to the inefficient combustion of saturated aliphatic and aromatic hydrocarbons [4]. The absence of benzaldehyde and decrease in tolualdehyde emissions with E-10 are due to the less aromatic content in E-10 compared to neat gasoline.

3.1.2 Comparison of BTEX Emissions

In case of E-10, benzene, toluene and p,m-xylene emissions decrease by 79.6, 28 and 31% respectively however, ethyl benzene and o-xylene increase by 41% and 11.8% respectively, as shown in Fig. 3(b). Consequently, there is an over all decrease of about 27% in BTEX emissions with E-10 as compared to E-0. This decrease in benzene and toluene in case of ethanol-containing fuel is also reported by other literature [9].

The reduction in BTEX components is due to the oxygen enrichment in ethanol, which is responsible for promoting the combustion rate in the combustion chamber, and hence for the oxidation of BTEX species in case of E-10, compared with E-0. According to Reuter, et. al. [17], benzene emissions were reduced by more than 10% with the use of oxygenated fuels like gasohol. Moreover, properties like lower boiling point, higher RON, faster flame propagation speed, and simple chemical structure of ethanol help E-10 in the quicker development of temperature. This quickly developed temperature is prone for decomposing the BTEX species especially, benzene, toluene and xylenes decrease significantly at higher temperature [18-20].
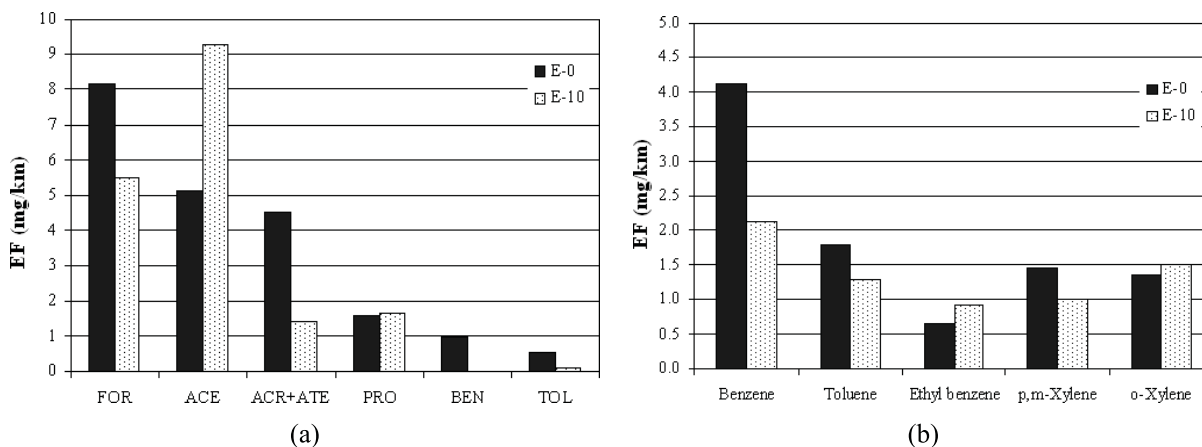

FIG. 3. COMPARISON OF (A) CARBONYLS AND (B) BTEX EMISSIONS IN TRANSIENT MODE

3.2   Steady-State Mode

3.2.1   Comparison of Carbonyls Emissions

Fig. 4(a) presents the emission of carbonyls at three different modes of 40, 80, and 120 km/hr. For all the steady-state modes of the vehicle, formaldehyde and acetaldehyde are the predominant components with E-0 and E-10, respectively. Once again, formaldehyde and acetaldehyde are the most abundant compounds in the total carbonyls, and their relative contributions are higher as compared to other components in both of the fuels.

Formaldehyde, acrolein + acetone, and propionaldehyde emissions are lower in case of E-10 as compared to E-0 for all the modes except mode 3 (120 km/hr) at which acrolein + acetone pollutants are higher with E-10 fuel. Acetaldehyde emissions are higher in case of E-10 relative to E-0 for all the steady-state modes of operation. As far as aromatic aldehydes are concerned, a few traces of benzaldehyde can be observed at modes 2 and 3, while tolualdehyde is found only at mode 3 with E-0. However, there is no evidence of such pollutants in case of E-10 fuel.

The reasons for lower formaldehyde, acrolein + acetone, and propionaldehyde emissions; absence of aromatic aldehydes; and higher acetaldehyde emissions with E-10 relative to E-0 are same as discussed earlier in
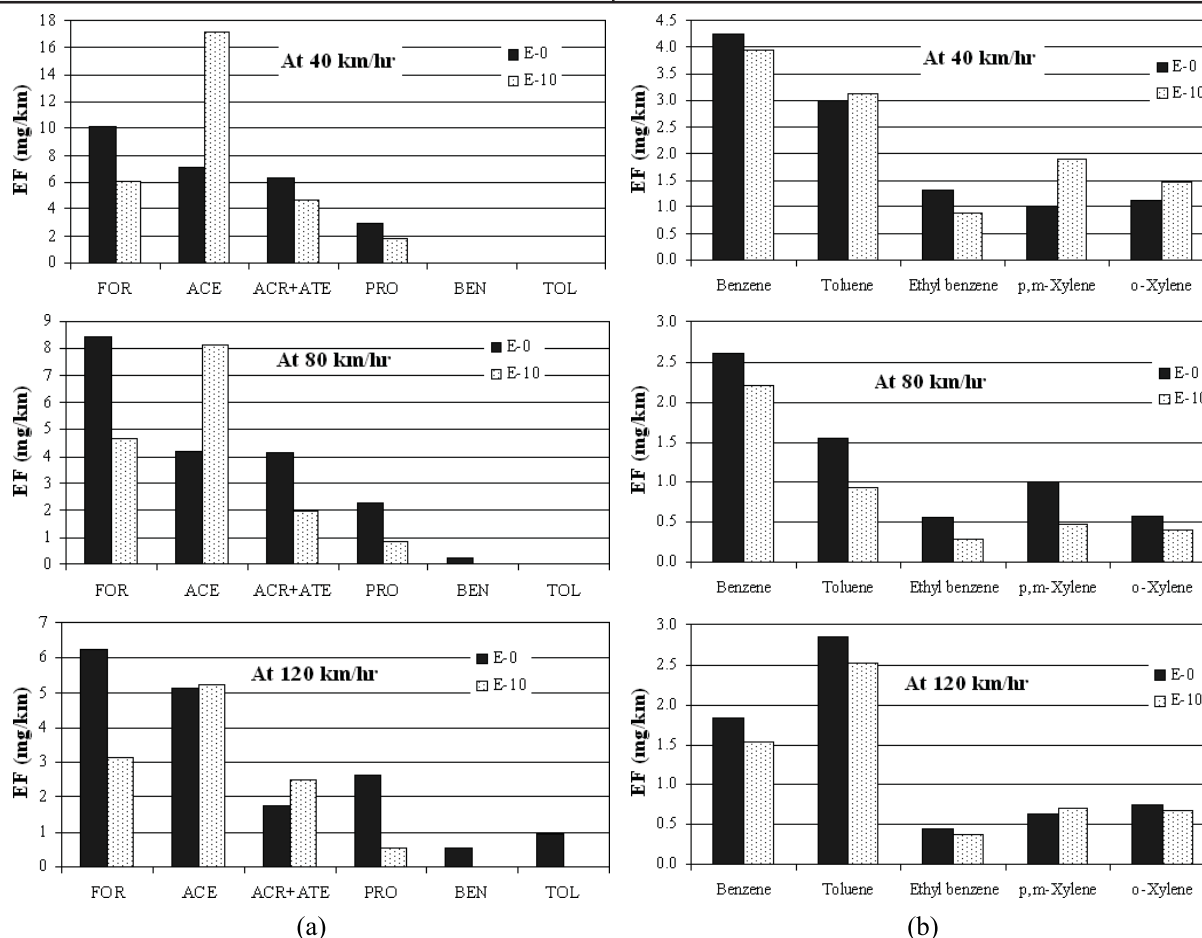

FIG. 4. COMPARISON OF (A) CARBONYLS AND (B) BTEX EMISSIONS AT STEADY-STATE MODES

Section 3.1.1. However, the increase in acroleine + acetone emissions at mode 3 may be due to the acrolein which might have come from the oxidation of glycerol and other residues of biofuels. So, probably this is the mode (speed level) of the vehicle, which is prone for the oxidation of glycerol, and hence for the increase in such emissions.

A strong correlation exits between formaldehyde and vehicle speed for both the fuels indicating that formaldehyde decreases as the speed progresses as shown in Fig. 4(a). This decrease in formaldehyde emissions at higher speeds is attributed to the improved combustion of the fuels at higher speeds relative to lower speed at which over-lean mixture area is increased, and thus combustion is retarded.

3.2.2 Comparison of BTEX Emissions

It is obvious from Fig. 4(b) that among the BTEX species, benzene is the predominant component at first two modes, whereas toluene is the most abundant component at the last mode for both the fuels. The emissions of benzene, toluene, and ethyl benzene decrease in case of E-10 relative to E-0, except mode 1 at which toluene is higher with E-10. However, p,m-xylene and o-xylene exhibit haphazard emission trends for different modes. The BTEX emissions decrease by 31.7 and 10.8% at mode 2 and 3 respectively; however, these pollutants increase by 6% at mode 1 with E-10 as compared to E-0. This decrease in BTEX emissions with E-10 relative to E-0 further strengthens the argument that combustion efficiency is increased in case of oxygenated fuel, particularly at higher speeds. Therefore, temperature is increased in the combustion chamber, which is prone for the decomposition of the BTEX components. The reason for higher BTEX emissions with E-10 compared to E-0 at mode 1 is the higher emissions of toluene and xylenes at this mode.

Again a correlation exists but this time between speed and Total BTEX emissions as shown in Fig. 5. Total BTEX emissions show their maxima at lower speed mode and minima at medium speed mode for both the fuels. This is attributed to the lower combustion temperature at lower engine speed of the vehicle because of the inefficient combustion caused by the development of over-lean mixture area in the combustion chamber, as mentioned above in Section 3.2.1. The over-lean mixture area results in incomplete combustion of the fuels.

3.3 Specific Reactivity of Pollutants

SR is the ratio of mg (milligram) O_3 (ozone) potential per milligram NMOG (Non-Methane Organic Gases) emanated from the exhaust, and can be evaluated as under [21]:

$$SR = \sum (NMOG_i \bullet MIR_i) / \sum NMOG_i \qquad (1)$$

The subscript i represents the certain pollutant emitted from the vehicle; NMOG is the sum of non-methane hydrocarbons and oxygenates, including carbonyls and BTEX species; and MIR is the maximum incremental reactivity. Carter and Lowi [21] performed air modeling based on ozone forming reactivates of species and gave the MIR factors as index for ozone formation, which are listed in Appendix, and indicate the maximum increase in ozone formation.

Fig. 6 presents the comparative SR of emissions from the vehicle in both transient as well as steady-state modes.
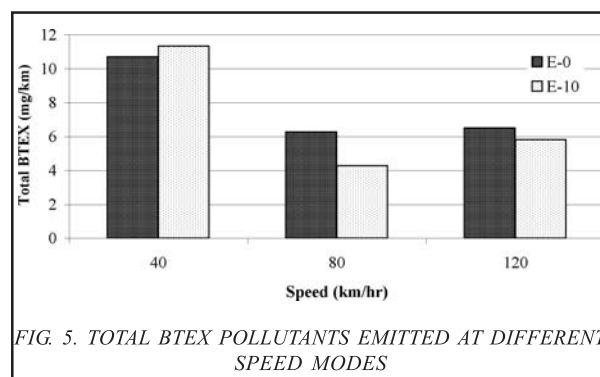

FIG. 5. TOTAL BTEX POLLUTANTS EMITTED AT DIFFERENT SPEED MODES

For the steady-state mode, mean value of the pollutants measured at three different modes was taken. It is clearly displayed that the pollutants in the transient mode of operation have higher SR as compared to those emitted in steady mode. E-10 exhibits the lower SR as compared to E-0. This leads to an advantage of E-10 over E-0 due to its lower contribution to ozone formation for both transient as well as steady-state modes.

4. CONCLUSIONS

Carbonyls and BTEX emissions were investigated from the tailpipe of an SI vehicle fuelled on gasoline and

FIG. 6. SPECIFIC REACTIVITY OF EMISSIONS IN TRANSIENT AND STEADY MODES

APPENDIX: MIR VALUES FOR CARBONYLS AND BTEX EMISSIONS [21]

BTEX	MIR	Carbonyls	MIR
Benzene	0.42	Formaldehyde	7.15
Toluene	2.73	Acetaldehyde	5.52
Ethyl Benzene	2.70	Acrolein+Acetone	6.77*,0.56**
p,m-Xylene	7.64	Propionaldehyde	6.53
o-Xylene	6.46	Benzaldehyde	-0.56
		Tolualdehyde	-0.56
*6.77 is for acrolein and *0.56 is for acetone			

ethanol- gasoline blend, and operated without TWC. In the transient mode of operation, formaldehyde was the predominant component of carbonyls in case of E-0, whose RC was above 55%. In case of E-10, acetaldehyde was the most abundant carbonyl compound exhibiting a RC of more than 58%. Formaldehyde, acrolein + acetone, and tolualdehyde were decreased but, acetaldehyde emissions were increased with E-10 compared to E-0. Benzene, toluene and p,m-xylene emissions were decreased whereas, ethyl benzene and o-xylene were increased with E-10. Consequently, there was an over all abatement of 27% in BTEX pollutants with E-10, relative to E-0.

During the steady-state modes of operation, formaldehyde and acetaldehyde were the predominant components for E-0 and E-10, respectively. A strong correlation was revealed between formaldehyde and vehicle speed for both the fuels, and formaldehyde was decreased as the speed increased. Formaldehyde, acrolein + acetone, and propionaldehyde emissions were lower, but acetaldehyde pollutants were higher with E-10 as compared to E-0. Aromatic aldehydes were absent in case of E-10 fuel. Among the BTEX species, benzene and toluene were the most abundant components from both the fuels. The emissions of benzene and ethyl benzene were lower in case of E-10 relative to E-0, for all the modes of operation. The BTEX emissions were decreased at mode 2 and 3 however, increased at mode 1 with E-10 as compared to E-0. Total BTEX emissions displayed their maxima at lower speed mode and minima at medium speed mode for both the fuels.

The ozone specific reactivity of the pollutants was higher for transient mode as compared to steady-state mode for both the fuels. Relative to E-0, E-10 revealed lower SR during both transient as well as steady-state modes.
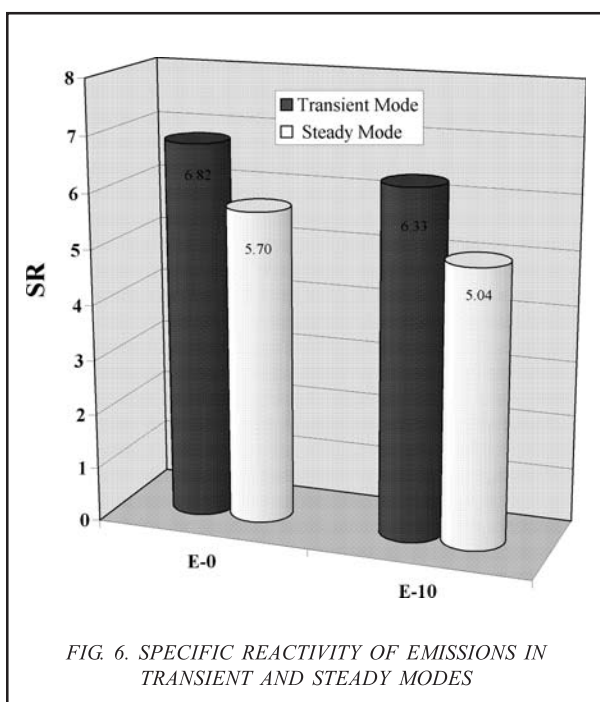
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